Homogeneously Alloyed $CdSe_{1-x}S_x$ QDs ($0 \le x \le 1$): an Efficient Synthesis for Full Optical Tunability

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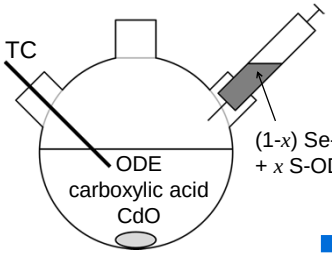
Introduction

In the field of fluorescent semiconductor quantum dots (QDs), alloyed QDs open up new possibilities and opportunities. Indeed, these systems allow to tune the optical properties of the nanocrystals without changing their size. This is of particular interest for the integration of the QDs in devices such as LEDs or for their use as biological labels. We recently developed a novel method for the synthesis of CdSe and ZnSe binary QDs in colloidal solutions that is fast and highly efficient.[1] This method is based on a heterogeneous Se-ODE precursor consisting of a simple dispersion of Se powder (200 mesh) in octadecene (ODE) and showing very high reactivity towards Cd precursor. In this contribution we will demonstrate that this method can be extended to the synthesis of $CdSe_{1-x}S_x$ homogeneously alloyed QDs ($0 \le x \le 1$).[2]

Synthesis

heterogeneous Se-ODE

homogeneous S-ODE

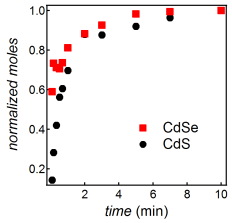


- ambient condition (air)
- T = 260°C
- 5 min reaction
- nearly full yield
- high solid load

➡ allow large production

Se-ODE vs. S-ODE

synthesis of pure CdSe and CdS (x = 0 and 1)

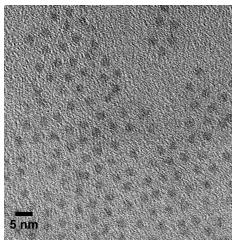


equivalent reactivity of both Se and S precursors towards Cd

→ prerequisite for homogeneous alloying

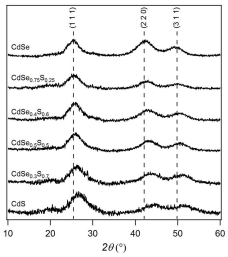
Structure and morphology

TEM of $CdSe_{0.5}S_{0.5}$ QDs



Ø ~ 3 nm with myristic acid
size can be tuned with the lenght of the carboxilic acid

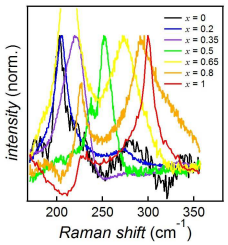
XRD vs. composition



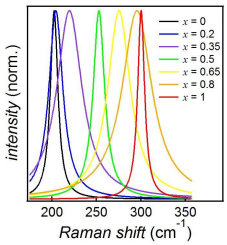
zincblende structure
lattice parameter shifts linearly with composition

Raman analysis

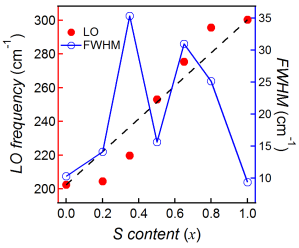
experimental spectra



fitted LO bands



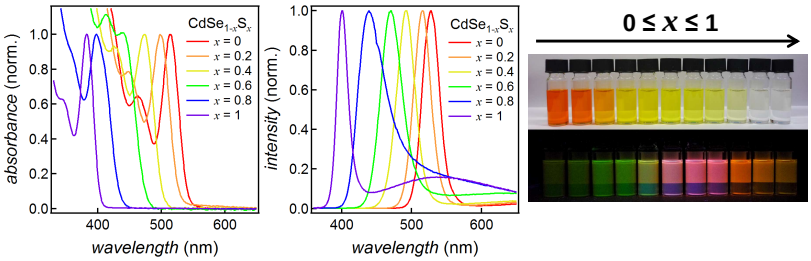
longitudinal optical (LO) phonon frequency



- LO frequency shifts with composition
- narrow FWHM for x = 0, 0.5 and 1
- large FWHM for uneven compositions

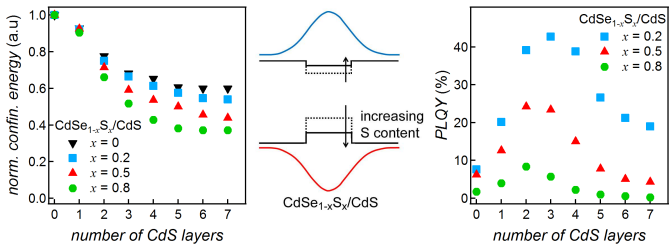
➡ homogeneous alloying

Optical properties



➡ efficient tuning of the band gap with the composition

$CdSe_{1-x}S_x$/CdS core-shell QDs



Confinement energy decreases faster for higher S content when growing CdS layers due to smaller band offset
→ increase of the PLQY is less pronounced

Conclusion

We developed a new, fast and phosphine-free method for the synthesis of alloyed $CdSe_{1-x}S_x$ QDs with $0 \le x \le 1$. The equivalent reactivity of the heterogeneous Se-ODE and homogeneous S-ODE precursors towards the Cd precursor leads to a homogeneous alloying as demonstrated by Raman spectroscopy. The optical properties of the QDs could easily be tuned by changing their composition. For $CdSe_{1-x}S_x$/CdS core-shell QDs, the efficiency of the shell in confining the exciton in the core decreases when increasing the S content due to a smaller interfacial band offset.

References

(1) S. Flamée, Z. Hens *et al.*, *submitted*
(2) T. Aubert, Z. Hens *et al.*, *submitted*

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